Comparative removal of emerging contaminants from aqueous solution by adsorption on an activated carbon

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Abstract

Batch sorption experiments were performed to study the adsorption of six emerging pollutants from aqueous solutions using a commercial granular activated carbon as adsorbent. Caffeine, clofibric acid, diclofenac, gallic acid, ibuprofen and salicylic acid were selected as representative contaminants. The activated carbon was characterized by nitrogen adsorption at 77 K, and through the determination of point of zero charge. The effects of several operational parameters, such as pH, initial concentration of organic molecules, mass of adsorbent and contact time, on the sorption behavior were evaluated. The contact time to attain equilibrium for maximum adsorption was found to be 40 min. The kinetic data were correlated to several adsorption models and the adsorption mechanism found to follow pseudo-second-order and intraparticle-diffusion models, with external mass transfer predominating in the first 15 min of the experiment. The equilibrium adsorption data were

analyzed using the Freundlich, Langmuir and Toth isotherm equation models. The similar chemical structure and molecular weight of the organic pollutants studied make the adsorption capacity of the activated carbon used very similar for all the molecules.

Keywords: adsorption, activated carbon, emerging water pollutants, pharmaceutical pollutants.

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1. Introduction

Although research studies in the pollutants field have tended to focus on the problem of priority pollutants, attention paid to emerging unregulated compounds increases continuously. Future regulation for emerging pollutants depends on the results obtained from the research on their potential health effects and from the monitoring of data regarding their occurrence [1]. Examples of this kind of compounds are pharmaceuticals, steroids and hormones, surfactants, flame retardants, industrial additives and agents, and gasoline additives [1]. Many undesired effects of these contaminants and their degradation products have been summarized in the literature [2]. Some of these contaminants do not even need to be persistent in the environment to cause negative effects, since their high transformation rate is compensated by their continuous introduction into the aquatic medium [3].

Several recent studies on the removal of emerging pollutants from water have revealed that coagulation-flocculation processes are generally not suitable [4,5]. Conventional biological treatment of these pollutants in wastewater treatment plants is time-consuming and not always effective, since not the whole range of the emerging pollutants can be removed this way [6]. Alternative technologies, such as adsorption on porous materials, use of membrane reactors, advanced oxidation processes and catalytic wet oxidation, have been found to be effective in removing several organic compounds from water [2].

Adsorption is an attractive method for the removal of contaminants from effluents since, if the adsorption system is designed correctly, it will provide a high-quality treated effluent which can be suitable for reuse. In comparison with other processes for the treatment of polluted aqueous effluents, the adsorption process allows flexibility in terms of both design and operation. Additionally, as the adsorption is sometimes reversible, the sorbent can be regenerated, thereby resulting in significant cost savings. Regarding the choice of the adsorbent, activated carbons are usually the first option in water treatment applications where

competitive adsorption is not important. Alternative adsorbents can be zeolites [7-9], metal organic frameworks [10-12] and clays [9,13-18], amongst others.

The adsorption of emerging pollutants on activated carbons has been studied by several authors over the past decade [19-27]. Thus, removal of clofibric acid from aqueous solution using cork-based activated carbons was studied by Mestre et al. [20]. These authors noted the importance of pH for clofibric acid adsorption and explained their findings on the basis of the solvation energies of undissociated and dissociated clofibric acid species rather than the chemical properties of the adsorbent. Similarly, the adsorption of caffeine, diclofenac, ibuprofen, carbamazepine and tetracycline on various activated carbons was investigated by Álvarez-Torrellas et al. [24,27], who linked their experimental adsorption results to the textural and chemical surface properties of the adsorbents. Rakic et al. [26] evaluated the ability of three commercial activated carbons to adsorb salicylic acid, acetylsalicylic acid, atenolol and diclofenac. The maximal adsorption capacities were correlated to the textural properties of the activated carbons while the acid/base features of the adsorbents and the molecular structures of the adsorbate molecules also played a role. Other carbon based materials as carbon nanotubes, chitosan, biochars, graphene, polymers, among others, have also been evaluated as adsorbents for the removal of organic contaminants [28-32].

In light of the above, the present study was motivated to test the effectiveness of a commercial granular activated carbon for the removal by adsorption of emerging contaminants at a concentration of up to 15 mg/dm^3 in water, the maximum solubility limit of all organic molecules. Caffeine (CF), clofibric acid (CA), diclofenac (DF), gallic acid (GA), ibuprofen (IB) and salicylic acid (SA) were considered the representative contaminants. An additional objective of the work was to test various kinetic and equilibrium models for the adsorption of these emerging contaminants on the granular activated carbon in single-component systems.

2. Experimental procedure

2.1. Materials

The adsorbent used in this work was a granular acid-washed activated carbon, Hydrodalco 3000, kindly supplied by Cabot Corporation. This carbon is produced by steam activation of coal and is used in water purification because of its excellent adsorption capacity for organics.

The chemical reagents were purchased from Sigma-Aldrich and were all of high purity (>98-99%). The chemical structures of all organic molecules used as adsorbates are shown in Table 1. The main physicochemical characteristics of the adsorbates, obtained from the ChemIDplus advanced database, are also included in Table 1. The aqueous solutions used in adsorprtion experiments were prepared using ultrapure water obtained with a Milli-Q apparatus (Millipore).

2.2. Characterization techniques

The point of zero charge was determined using the salt addition method [33]. The initial pH values (pH_i) of aqueous solutions of NaCl 0.01 mol/dm^3 were adjusted to a pH range of 2–12 using either 0.1 mol/dm^3 HCl or 0.1 mol/dm^3 NaOH; then, 0.15 g of adsorbent was added to 50 cm^3 of solution. The suspensions were sealed and shaken for 48 h at ambient temperature; finally they were filtered and the pH (pH_f) of the supernatant was measured. The shift in pH, $\Delta pH = pH_i - pH_f$, was plotted against pH_i and the point of zero charge was determined as the intersection of the curve with the pH axis.

Nitrogen (>99.999%) adsorption measurements at 77 K were performed on a Micromeritics ASAP 2010 volumetric adsorption analyzer. In each run, about 0.1 g of activated carbon was used, after outgassing at 473 K for 16 h to remove any adsorbed

molecules. The adsorption-desorption isotherms were obtained over the relative pressure range from 0 to 1 to determine the surface area and the pore volume.

2.3. Adsorption procedure

The adsorption experiments were performed in batch mode. In kinetic tests, and considering previous works of our research group [34], 50 mg of adsorbent was added to glass tubes containing 10 cm^3 aqueous solutions of the pollutants at initial concentrations of 5, 10 and 15 mg/dm^3. The effect of the adsorbent dose was studied by varying the amount of the absorbent (25, 50 and 100 mg) which was added to the solutions with an initial pollutant concentration of 15 mg/cm^3. The effect of the pH on the adsorption was studied for the solutions that contained 15 mg/dm^3 pollutant and 50 mg adsorbent, for which the initial pH values were adjusted to 2.8 or 6.0 by proper addition of HCl or NaOH. After been agitated for a predetermined time in an orbital shaker at room temperature, the suspensions were filtered through 0.45 μm Durapore membrane filters. The concentration of the pollutant remaining in aqueous solution was measured using a Jasco V-730 UV-Vis spectrometer at the maximum absorption wavelength which was determined experimentally for each molecule (Table 1).

Various contaminant solutions with several concentrations were prepared to determine the equilibrium adsorption capacity of the activated carbon. In equilibrium tests, 50 mg of adsorbent was added to glass tubes containing 10 cm^3 of the contaminant solution at a concentration ranging from 0 to 500 mg/dm^3. The pH was adjusted to 6.0 and the suspensions were agitated in the shaker for 2 h. The rest of the procedure was similar to the one previously described for kinetic tests. The amount of organic pollutant adsorbed by the solid at equilibrium was calculated according to the Equation 1, where C_e (mg/dm^3) is the pollutant concentration at equilibrium. All adsorption experiments were performed at least twice and the results are reported as an average.

2.3.1. Theoretical approach: Adsorption kinetics

Two types of kinetic modeling approaches have been considered in the adsorption literature to describe the transport of adsorbates inside adsorbent particles [35]. The first type considers simple relationships between the adsorption performance and operating conditions. These models show how the mean adsorbent loading (q_t) changes with adsorption time. Models in this category include pseudo-first- and pseudo-second-order rate equations. The parameters obtained from these models are used for adsorbent screening procedures. The second approach is the use of phenomenological models that attempt to describe the physics of the process. Information about the mechanism of adsorption can be obtained from the second type of models using the kinetic experimental results and the equilibrium adsorption data.

In order to investigate the adsorption process for the organic molecules in this study, pseudo-first- and pseudo-second-order rate equations and the intra-particle diffusion model were used and applied to test the experimental data. The first-order rate equation, or the so-called Lagergren equation, can describe the initial phase in the adsorption process, although as adsorption progresses the adsorption data may deviate from the fitted curve. The second-order rate equation suggests that chemisorption is the rate-controlling mechanism. Moreover, it has frequently been employed to analyze adsorption data obtained from various experiments with several types of adsorbates and adsorbents, as reviewed by Liu and Shen [36].

The mass balance for the adsorbate in a batch liquid-phase system before attainment of adsorption equilibrium, assuming first- or second-order kinetics for the driving force term, is represented by Equations (1) and (2), respectively:

$$-\frac{dn}{dt} = -V\frac{dC}{dt} = k_1 \cdot V \cdot (C - C_e) \qquad (1)$$

$$-\frac{dn}{dt} = -V\frac{dC}{dt} = k_2 \cdot \frac{V^2}{m} \cdot (C - C_e)^2 \quad (2)$$

where n (mg) is the adsorbate mass in the liquid phase, V (dm^3) is the volume of solution, C (mg/dm^3) is the solute concentration, m (g) is the adsorbent mass, t (min) is the time of contact and k_1 (1/min) and k_2 (g/mg·min) are the adsorption kinetic constants for pseudo-first and pseudo-second order kinetics, respectively.

Integrating the Equations (1) and (2) between $t = 0$ and time t, and considering the initial conditions (for $t = 0$, $C = C_0$), the following equations are obtained:

$$\ln\left(\frac{C_t - C_e}{C_0 - C_e}\right) = -k_1 \cdot t \quad (3)$$

$$C_t = C_e + \frac{V \cdot m \cdot (C_0 - C_e)}{m + k_2 \cdot V \cdot (C_0 - C_e) \cdot t} \quad (4)$$

The relationship between the concentration (C) of the organic molecule in the aqueous solution and the amount (q) adsorbed on the solid, based on the concept that the organic molecule mass that is removed from the liquid phase is transferred to the solid, is given by the following equation:

$$q_t = \frac{V \cdot (C_0 - C_t)}{m} \quad (5)$$

where q is expressed in mg of adsorbate per g of adsorbent.

Taking into account Equation (5), Equations (3) and (4) become:

$$q_t = q_e \cdot (1 - \exp(-k_1 \cdot t)) \tag{6}$$

$$q_t = \frac{k_2 \cdot q_e^2 \cdot t}{1 + k_2 \cdot q_e \cdot t} \tag{7}$$

Equations (6) and (7) are widely used to interprete the experimental results obtained for aqueous pollutants [10,13,27,37-39]. The parameters in these equations are estimated by fitting the models to the experimental data by non-linear regression analysis.

Intraparticle diffusion is the rate-controlling step in the case of well-mixed solutions, for which the intraparticle diffusivity is constant and the uptake of adsorbate by the adsorbent is low compared to the total quantity of adsorbate present in the solution. The equation for the intraparticle diffusion model is represented by the Morris and Weber approach:

$$q_t = k_3 \cdot t^{0.5} \tag{8}$$

where k_3 (mg/g·min$^{0.5}$) is the intraparticle diffusion rate constant. This model has been applied for several adsorption systems [13,27,40-42].

Following with the physics description of the adsorption process, the external mass transfer or boundary layer effects can be analyze from a model which assumes that the surface concentration of the adsorbate is negligible at time t = 0. If the system is well-stirred, the concentrations of adsorbate on the adsorbent surface and in the liquid phase must be uniform. The external mass-transfer coefficient can be obtained from the Furusawa and Smith model [43]:

$$\frac{C_t}{C_0} = \frac{1}{1 + m_S \cdot k_T} + \frac{m_S \cdot k_T}{1 + m_S \cdot k_T} \exp\left(- \frac{1 + m_S \cdot k_T}{m_S \cdot k_T} \cdot k_{FS} \cdot \frac{m}{V} S_{ext} \cdot t \right) \tag{9}$$

where k_T (dm^3/g) is the equilibrium constant in the Toth isotherm (see Equation 13), m_S (g/dm^3) is the concentration of adsorbent, S_{ext} (m^2/g) is the external specific surface area and k_{FS} (cm/s) is the external mass-transfer coefficient.

The effective diffusion coefficient for the adsorption process in porous adsorbents can be determined from the fractional approach to the equilibrium, *F(t)* [44]:

$$\frac{C_0 - C_t}{C_0 - C_e} = F(t) = \left[1 - \exp\left(-\frac{\pi^2 \cdot D \cdot t}{r^2}\right)\right]^{0.5} \tag{10}$$

where D (m^2/s) is the intraparticle-diffusion coefficient and r (m) is the particle size radius assuming a spherical geometry.

2.3.2. Theoretical approach: Adsorption models

The Freundlich isotherm model is an empirical equation employed to describe the adsorption processes in heterogeneous adsorbents, especially for organic compounds and highly interactive species on activated carbon [45]. The Freundlich isotherm equation can be expressed as follows:

$$q_e = k_F \cdot C_e^{\,1/m_F} \tag{11}$$

where k_F and m_F are empirical constants that indicate adsorption magnitude and effectiveness.

The well-known Langmuir model was developed for monolayer coverage of homogeneous adsorbent surfaces [45]:

$$q_e = \frac{k_L \cdot q_L \cdot C_e}{1 + k_L \cdot C_e} \tag{12}$$

where k_L (dm^3/mg) and q_L (mg/g) are the equilibrium adsorption constant related to the affinity of binding sites, and the monolayer adsorption capacity, respectively.

Finally, the Toth model is an improvement of the Freundlich and Langmuir equations. The equation describes the adsorption behavior of various adsorbates under sub-critical conditions.

$$q_e = \frac{k_T \cdot q_T \cdot C_e}{[1 + (k_T \cdot C_e)^{m_T}]^{1/m_T}} \tag{13}$$

where q_T (mg/g), k_T (dm^3/mg) and m_T are the Toth constants.

3. Results and discussion

3.1. Characterization of the adsorbent

The nitrogen adsorption isotherm for the activated carbon studied herein was of type II in the Brunauer, Deming, Deming and Teller (BDDT) classification [46], and the hysteresis loop was of type IIIb in the Rouquerol et al. classification [47]. The BET specific surface area was found to be 578 m^2/g and the total pore volume obtained at a relative pressure of 0.98 was 0.564 cm^3/g. The external specific surface area and micropore volume, estimated using the *t*-plot method [46], were 159 m^2/g and 0.206 cm^3/g, respectively. These results indicate that the activated carbon used as adsorbent is a mesoporous material.

3.2. Adsorption experiments

Several factors, such as the solution pH, the adsorbent dose and the initial concentration of the adsorbate, amongst others, can play important roles in the adsorption process. The pH can simultaneously affect the surface charge of the adsorbent, the degree of ionization of functional groups of the adsorbate, and the adsorption mechanism [20]. Thus, the effect of pH on the removal of organic molecules by the activated carbon was studied to gain further insight into the adsorption process. To that end, the effect of pH on the adsorption was determined at values of 2.8 and 6, with a CA, GA and SA feed concentration of 15 mg/dm^3 and an adsorbent dose of 50 mg. The results obtained are shown in Figure 1. It can be observed that the percentage organic molecule uptake increases with contact time until an almost constant value is reached. At this point the amount of adsorbate being removed from aqueous solution by the activated carbon is in a state of dynamic equilibrium with the amount of organic molecule desorbed from the activated carbon. It can also be seen that CA is adsorbed to a greater extent, followed by GA and SA. Considering both pHs, no appreciable differences were observed for the amount of salicylic acid retained on the surface of the activated carbon. For the other two molecules, the amount adsorbed increases by between 19% and 23% when the pH is below pH_{pzc}.

The adsorbent dose is an important parameter that strongly influences the adsorption process by affecting the adsorption capacity of the adsorbent. Various doses of between 25 and 100 mg activated carbon were investigated in this study, and the results are summarized in Figures 2 and 3. It can be seen that the equilibrium adsorption capacity decreased with increasing amount of adsorbent dose. This phenomenon may be explained as a result of particle interactions such as partial overlapping or aggregation of adsorption sites, thus causing a decrease in the effective adsorbent surface area available to organic molecules and an increase in the diffusion path length.

The initial organic molecule concentration and contact time is another controlling parameter that strongly influences the adsorption process. As such, a plot of equilibrium adsorption capacity versus contact time for various initial organic molecule concentrations of between 5 and 15 mg/dm^3 is also shown in Figures 2 and 3. Experiments were performed with a fixed adsorbent dose of 50 mg, at a fixed temperature at pH 6, and shaken until equilibrium was achieved. The adsorption capacity for the activated carbon increased from 0.60 to 3.07 mg/g in the case of CA, GA and SA, and from 0.18 to 3.14 mg/g in the case of CF, DF and IB, as the initial concentration of the organic molecule increased from 5 to 15 mg/dm^3. This trend can be explained from the perspective of a concentration gradient, which acts as the driving force to overcome the mass transfer resistance of the organic molecule between the aqueous and solid phases. Under our experimental conditions, a higher initial concentration provides a higher driving force for mass transfer of organic molecules, thus leading to a higher adsorption capacity until saturation. The adsorption of organic molecules showed a two-stage behavior: a very rapid initial adsorption over a few minutes (up to 40 min), followed by a longer period of much slower uptake. During the initial stage, a large number of active sites on the adsorbent and the interaction between the surface of the adsorbent and the organic molecule enhanced the adsorption. The subsequent lower adsorption may be due to repulsive forces between the organic molecules adsorbed on the adsorbent and those in the bulk phase. More information about this process can be obtained when considering the diffusion mechanism (see the next section). Representative plots of adsorption capacity versus contact time are provided in Figure 4.

3.3. Kinetic models

The results relating to kinetic behavior (see Figures 2 and 3, and Tables 2 and 3) reveal that the adsorption of organic molecules can best be described as a pseudo-second-order

linear reaction. An analysis of the k_2 values (see Table 3) shows the k_2 values for the acids are greater than those for pharmaceutical molecules (CF, DF and IB). Thus, the adsorption rate of acids on the activated carbon is faster than that for the pharmaceutical molecules. The results also show that the adsorption rate constant decreases as the initial organic molecule concentration increases from 5 to 15 mg/dm^3 and as the adsorbent mass increases from 25 to 100 mg. An increase in adsorbent mass increases the adsorption rate constant as a result of the higher number of active sites available for adsorption. This process was described by Chu [48], who indicated that, at low initial adsorbate concentration, the adsorbate will bind preferentially to high energy sites. The sites of higher energy are usually taken up by adsorbate molecules first, with sites of lower energy progressively filled as adsorbate loading is increased. It therefore follows that the adsorbate bond in low adsorbate loading situations will result in faster reaction kinetics. Adsorption on lower energy sites, as in the case of high loading, will subsequently result in a decrease in the k_2 value.

Mass-transfer processes in porous solids are always complicated because of molecular sieve and diffusional effects. Diffusion takes place when the dimensions of the adsorbate molecules are smaller than the pore diameter. However, internal diffusion can be neglected for nonporous, macroporous and mesoporous solids, thus meaning that control is determined by external diffusion processes. For brevity reasons the external mass-transfer coefficients were determined using the Furusawa–Smith method for CF, DF and IB only, and the results are shown in Figure 5 and Table 5. An increase in the initial organic molecule concentration decreases the initial rate of adsorption, as also reported by McKay et al. [49]. This behavior can be attributed to interactions between solute molecules in solution and to increased competition for available adsorption sites. An increase in adsorbent mass decreases the resistance to external mass-transfer and increases the external mass-transfer coefficient.

Plots of the Morris–Weber relationship for sorption of the organic molecules by the activated carbon, at an initial concentration of 10 mg/dm^3, are shown in Figure 6. In light of this figure, it can be seen that the intra-particle diffusion of organic molecules within the activated carbon occurs in two stages [40]. The first linear portion includes the adsorption period from 0 to 16 min and represents the external mass transfer or boundary layer diffusion and the rapid distribution of molecules onto the outer surface of the activated carbon. The second linear portion corresponds to the adsorption period from 25 to 90 min, which represents the intra-particle diffusion and binding of molecules into the internal active sites of the activated carbon. The slope of the linear portion (see Table 4) indicates the rate of the adsorption process, which decreases with contact time. As the organic molecule diffuses into the inner structure of the activated carbon, the porosity available for diffusion decreases. The calculated effective diffusion coefficients are summarized in Table 6. The relatively low value of D (10^{-5} L/s) also indicates the low role of intraparticle diffusion in the adsorption of organic molecules on the mesoporous activated carbon. It is clear from the data that, as the mass of adsorbent increases from 25 to 100 mg, the effective diffusion coefficients also increase. Various explanations for this have been proposed by several authors when studying dye adsorption [50-52].

3.4. Adsorption isotherms

The most common representation of adsorbate concentration and quantity of material adsorbed is the adsorption isotherm. The equilibrium adsorption isotherm is fundamental for describing the interactive behavior between solutes and adsorbents, and is the basic requirement in the design of adsorption systems [53]. In this study, the Freundlich, Langmuir and Toth isotherm equations were used to model the experimental data for SA, CF, DF and IB. Adsorption isotherms for the retention of these organic molecules by the activated carbon

are presented in Figure 7. These isotherms show a characteristic L-type behavior according to the Giles classification, thereby representing a system in which the adsorbate is strongly attracted by the adsorbent [54]. The calculated constants for the isotherm equations, as estimated by non-linear regression, are presented in Table 5. Although all models describe the experimental results well, taking into consideration the parameters obtained, the Toth equation gives better agreement between the theoretical and experimental data than the others. This finding is consistent with other studies which demonstrate that the Toth model correctly describes the adsorption of organic molecules on activated carbons [39].

3.5. Mechanism of emerging contaminants adsorption on the activated carbon

It has been reported several possible interactions that can explain the adsorption of organic contaminants on adsorbents: hydrophobic interactions, π-π stacking interactions, van der Waals forces, electrostatic interactions and hydrogen bonding interactions that can act individually or simultaneously [28,29]

The results of adsorption suggest that the commercial adsorbent show very strong adsorption affinity for the organic compounds considered in this study. The attractive or repulsive interactions between the adsorbent surface and the adsorbate are affected by the chemical characteristics of the adsorbents. The point of zero charge (pH_{pzc}) is an important chemical characteristic of adsorbents as it determines the net charge of the adsorbent surface in the solution. Adsorbents have a net negative charge on their surface when the equilibrium pH of the solution is higher than their pH_{pzc}, whereas they have a net positive charge when the equilibrium pH in solution is lower than their pH_{pzc}. The pH_{pzc} value determined for the activated carbon was 4.5. Therefore, the adsorbent has negative charge as the equilibrium pH of 6 is higher than the pH_{pzc} value, thus meaning that electrostatic attractions between the negatively charged surface and the positively charged organic molecules could favor

adsorption. However, the experimental results also show that there is an interaction between the surface of the activated carbon and the adsorbates at pH 2.8, thus suggesting that the effect of van der Waals and π-π interactions cannot be neglected. This type of interactions mainly occurred between the π-electron-depleted aromatic rings (also regions) and the π-electron-rich regions (or aromatic rings) of the adsorbate and the adsorbent. The role donor/acceptor is not fixed [55].

The molecular size and shape of organic contaminants have also been reported that determine the availability of the adsorption sites on adsorbents. In general, larger molecules have higher adsorption energies, and thus larger differences in molecular size result in better separation in a system with mixed chemicals [56,57]. The rate of organic chemical diffusion is also dependent on molecular size and shape. The organic molecules used in this work all have a fairly similar chemical structure between them, as well as a similar molecular weight. Therefore, the behavior in the adsorption by the activated carbon will also be very similar

5. Conclusions

In the present study the ability of a commercial granular activated carbon to remove six emerging pollutants from aqueous solutions was evaluated and compared. The results obtained show that the adsorbent shows a similar adsorption capacity for all adsorbates. These results can be explained considering the limited surface chemistry of this type of material.

The kinetic study revealed that stirring for 40 min is sufficient for the organic molecule/activated carbon system to reach equilibrium. The experimental kinetic data were fitted to pseudo-first- and pseudo-second-order rate equations and the intra-particle diffusion model. The experimental results can best be described as a pseudo-second-order linear reaction. The low value of the calculated effective diffusion coefficients indicates the limited

role of intraparticle diffusion in the adsorption of the emerging contaminants on the mesoporous activated carbon.

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Captions

Table 1. General characteristics of the adsorbates (available from: http://chem.nlm.nih.gov/chemidplus/).

Table 2. Pseudo-first-order parameters for the adsorption of organic molecules by the activated carbon. T = 298 K and pH 6.

Table 3. Pseudo-second-order parameters for the adsorption of organic molecules by the activated carbon. T = 298 K and pH 6.

Table 4. External mass transfer for the adsorption of organic molecules by the activated carbon. T = 298 K and pH = 6.

Table 5. Intraparticle rate parameters for the adsorption of organic molecules by the activated carbon. T = 298 K and pH 6.

Table 6. Effective diffusion coefficients for the adsorption of organic molecules by the activated carbon. T = 298 K and pH 6.

Table 7. Freundlich, Langmuir and Toth parameters for the adsorption of organic molecules by the activated carbon. Equilibrium time = 2 h, T = 298 K, pH 6.

Figure 1. Kinetic adsorption data for clofibric (CA), gallic (GA) and salicylic (SA) acids on activated carbon considering two adsorption pHs (2.8 and 6). T = 298 K, 15 mg/dm^3 and 50 mg.

Figure 2. Kinetic adsorption data for clofibric, gallic and salicylic acids on the activated carbon considering various acid concentrations and adsorbent amounts. T = 298 K, pH 6. The lines represent a pseudo-first-order model (—) and a pseudo-second-order model (---).

Figure 3. Kinetic adsorption data for caffeine, diclofenac and ibuprofen on the activated carbon considering various acid concentrations and adsorbent amounts. T = 298 K, pH 6. The lines represent a pseudo-first-order model (—) and a pseudo-second-order model (---).

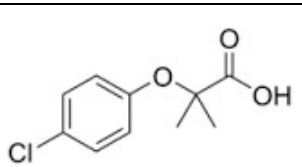
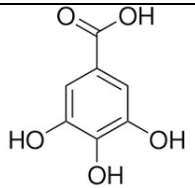
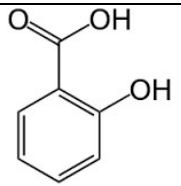
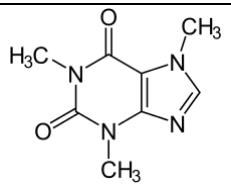
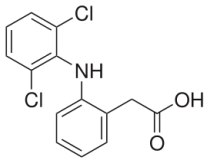
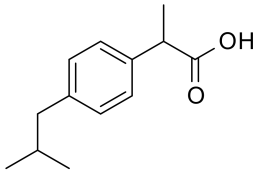
Figure 4. Kinetic adsorption data for organic molecules on the activated carbon considering 15 mg/dm^3 of acid and 25 mg of adsorbent. T = 298 K, pH 6. The lines represent a pseudo-first-order model (—) and a pseudo-second-order model (---).

Figure 5. Furusawa–Smith model for the adsorption of organic molecules on the activated carbon. T = 298 K and pH 6.

Figure 6. Intraparticle diffusion model for the adsorption of organic molecules on the activated carbon. T = 298 K and pH 6.

Figure 7. Experimental (symbols) and model (lines) isotherms for the equilibrium adsorption of organic molecules on the activated carbon. Equilibrium time = 2 h, T = 298 K, pH 6.

Table 1. General characteristics of the adsorbates (available from: http://chem.nlm.nih.gov/chemidplus/).

Characteristic	Clofibric acid	Gallic acid	Salicylic acid	Caffeine	Diclofenac	Ibuprofen
Molecular structure						
IUPAC name	2-(4-Chlorophenoxy)-2-methylpropionic acid	3,4,5-Trihydroxybenzoic acid	2-Hydroxybenzoic acid	1,3,7-Trimethylpurine-2,6-dione	2-(2,6-Dichloranilino) phenylacetic acid	(*RS*)-2-(4-(2-Methylpropyl)phenyl) propanoic acid
Chemical formula	$C_{10}H_{11}ClO_3$	$C_7H_6O_5$	$C_7H_6O_3$	$C_8H_{10}N_4O_2$	$C_{14}H_{11}Cl_2NO_2$	$C_{13}H_{18}O_2$
Molecular weight	214.65	170.12	138.12	194.19	296.15	206.28
pK_a[1]	2.8	3.13	2.97	10.4	4.15	4.9
λ_{max} (nm)[2]	227	268	230	273	276	222

[1] K_a : dissociation constant at 20°C

[2] λ_{max} : maximum absorption wavelength, determined experimentally

Table 2. Pseudo-first-order parameters for the adsorption of organic molecules by the activated carbon. T = 298 K and pH 6.

	Clofibric acid	Gallic acid	Salicylic acid	Caffeine	Diclofenac	Ibuprofen
5 mg/dm^3 – 50 mg						
k_1 (1/min)	0.34	0.093	0.083	0.11	0.14	0.09
χ^2	0.033	0.10	0.11	0.0025	0.0017	0.0068
R	0.96	0.94	0.96	0.96	0.98	0.91
10 mg/dm^3 – 50 mg						
k_1 (1/min)	0.63	0.060	0.086	0.11	0.11	0.18
χ^2	0.083	0.15	0.12	0.04	0.04	0.067
R	0.97	0.98	0.992	0.98	0.98	0.96
15 mg/dm^3 – 50 mg						
k_1 (1/min)	0.18	0.12	0.097	0.12	0.095	0.061
χ^2	2.8	0.34	1.5	0.14	0.23	0.24
R	0.82	0.98	0.93	0.993	0.98	0.990
15 mg/dm^3 – 25 mg						
k_1 (1/min)	0.19	0.032	0.25	0.041	0.053	0.027
χ^2	4.0	0.36	4.0	0.97	0.32	1.27
R	0.84	0.997	0.93	0.98	0.994	0.98
15 mg/dm^3 – 100 mg						
k_1 (1/min)	0.26	0.14	0.26	0.14	0.20	0.18
χ^2	0.51	0.048	0.16	0.025	0.083	0.079
R	0.87	0.991	0.97	0.995	0.98	0.990

Table 3. Pseudo-second-order parameters for the adsorption of organic molecules by the activated carbon. T = 298 K and pH 6.

	Clofibric acid	Gallic acid	Salicylic acid	Caffeine	Diclofenac	Ibuprofen
5 mg/dm^3 – 50 mg						
k_2 (g/mg·min)	1.2	0.21	0.13	0.93	1.03	0.69
χ^2	0.024	0.12	0.13	0.0020	0.0027	0.005
R	0.97	0.94	0.95	0.980	0.97	0.96
10 mg/dm^3 – 50 mg						
k_2 (g/mg·min)	1.3	0.047	0.062	0.18	0.17	0.36
χ^2	0.023	0.35	0.22	0.046	0.025	0.031
R	0.992	0.97	0.98	0.98	0.993	0.98
15 mg/dm^3 – 50 mg						
k_2 (g/mg·min)	0.11	0.071	0.047	0.070	0.048	0.029
χ^2	1.3	0.63	0.59	0.61	0.28	0.59
R	0.92	0.97	0.97	0.98	0.991	0.98
15 mg/dm^3 – 25 mg						
k_2 (g/mg·min)	0.10	0.0092	0.10	0.011	0.013	0.0077
χ^2	1.8	2.7	2.3	2.32	2.14	3.61
R	0.93	0.98	0.96	0.98	0.98	0.98
15 mg/dm^3 – 100 mg						
k_2 (g/mg·min)	0.37	0.17	0.37	0.15	0.24	0.21
χ^2	0.18	0.039	0.073	0.050	0.063	0.20
R	0.95	0.993	0.98	0.993	0.98	0.97

Table 4. External mass transfer for the adsorption of organic molecules by the activated carbon. T = 298 K and pH = 6.

	Caffeine	Diclofenac	Ibuprofen
5 mg/dm^3 – 50 mg			
k_{FS} (cm/s)	2.17 10^{-7}	2.34 10^{-7}	29.5 10^{-7}
χ^2	0.20	0.30	0.48
R	0.91	0.86	0.76
10 mg/dm^3 – 50 mg			
k_{FS} (cm/s)	2.13 10^{-7}	1.87 10^{-7}	12.0 10^{-7}
χ^2	0.057	0.038	0.16
R	0.98	0.98	0.92
15 mg/dm^3 – 50 mg			
k_{FS} (cm/s)	1.51 10^{-7}	1.62 10^{-7}	1.63 10^{-7}
χ^2	0.035	0.038	0.035
R	0.98	0.98	0.991
15 mg/dm^3 – 25 mg			
k_{FS} (cm/s)	1.48 10^{-7}	1.34 10^{-7}	0.93 10^{-7}
χ^2	0.034	0.0085	0.048
R	0.991	0.997	0.98
15 mg/dm^3 – 100 mg			
k_{FS} (cm/s)	2.13 10^{-7}	1.97 10^{-7}	8.2 10^{-7}
χ^2	0.012	0.046	0.046
R	0.996	0.98	0.98

Table 5. Intraparticle rate parameters for the adsorption of organic molecules by the activated carbon. T = 298 K and pH 6.

	Clofibric acid	Gallic acid	Salicylic acid	Caffeine	Diclofenac	Ibuprofen
5 mg/dm^3 – 50 mg						
k'_3 (mg/g·min$^{0.5}$)	0.20	0.068	0.21	0.027	0.059	0.020
R	0.98	0.997	0.97	0.94	0.97	0.95
k''_3 (mg/g·min$^{0.5}$)	0.0061	0.052	0.037	0.0026	0.0049	0.0020
R	0.96	0.98	0.98	0.69	0.75	0.5
10 mg/dm^3 – 50 mg						
k'_3 (mg/g·min$^{0.5}$)	0.23	0.18	0.58	0.17	0.22	0.19
R	0.992	0.96	0.97	0.97	0.97	0.94
k''_3 (mg/g·min$^{0.5}$)	0.0023	0.076	0.044	0.019	0.035	0
R	0.96	0.93	0.94	0.97	0.98	0.48
15 mg/dm^3 – 50 mg						
k'_3 (mg/g·min$^{0.5}$)	0.14	0.23	0.43	0.70	0.58	0.46
R	0.96	0.95	0.94	0.98	0.97	0.95
k''_3 (mg/g·min$^{0.5}$)	0.13	0.032	0.21	0.0070	0.087	0.068
R	0.85	0.98	0.97	1	0.97	0.85
15 mg/dm^3 – 25 mg						
k'_3 (mg/g·min$^{0.5}$)	0.15	0.39	0.94	0.81	0.81	0.71
R	0.98	0.81	0.95	0.96	0.97	0.97
k''_3 (mg/g·min$^{0.5}$)	0.10	0.21	0.13	0.55	0.085	0.12
R	0.92	0.991	0.92	0.998	0.97	0.97
15 mg/dm^3 – 100 mg						
k'_3 (mg/g·min$^{0.5}$)	0.12	0.41	0.31	0.38	0.38	0.44
R	0.90	0.92	0.93	0.996	0.991	0.97
k''_3 (mg/g·min$^{0.5}$)	0.041	0.013	0.023	0.0081	0.00009	--
R	0.85	0.94	0.97	0.83	0.03	--

Table 6. Effective diffusion coefficients for the adsorption of organic molecules by the activated carbon. T = 298 K and pH 6.

	Caffeine	Diclofenac	Ibuprofen
5 mg/dm^3 – 50 mg			
D/r^2 (1/s)	2.24 10^{-5}	2.42 10^{-5}	1.76 10^{-5}
χ^2	0.12	0.19	0.20
R	0.97	0.95	0.95
10 mg/dm^3 – 50 mg			
D/r^2 (1/s)	3.03 10^{-5}	2.95 10^{-5}	5.44 10^{-5}
χ^2	0.16	0.11	0.075
R	0.97	0.98	0.98
15 mg/dm^3 – 50 mg			
D/r^2 (1/s)	3.60 10^{-5}	2.56 10^{-5}	1.89 10^{-5}
χ^2	0.27	0.17	0.30
R	0.95	0.96	0.94
15 mg/dm^3 – 25 mg			
D/r^2 (1/s)	1.40 10^{-5}	1.29 10^{-5}	1.05 10^{-5}
χ^2	0.36	0.41	0.43
R	0.93	0.89	0.91
15 mg/dm^3 – 100 mg			
D/r^2 (1/s)	4.49 10^{-5}	7.15 10^{-5}	5.47 10^{-5}
χ^2	0.13	0.11	0.24
R	0.97	0.97	0.94

Table 7. Freundlich, Langmuir and Toth parameters for the adsorption of organic molecules by the activated carbon. Equilibrium time = 2 h, T = 298 K, pH 6.

	Salicylic acid	Caffeine	Diclofenac	Ibuprofen
Freundlich				
q_F	7.3	12	5.0	9.5
m_F	3.7	2.4	2.4	4.2
χ^2	46	252	37	74
R	0.97	0.98	0.993	0.97
Langmuir				
q_L (mg/g)	33	88	64	34
k_L (dm^3/mg)	0.073	0.094	0.014	0.26
χ^2	152	58	115	143
R	0.92	0.996	0.98	0.95
Toth				
q_T (mg/g)	422	112	2557	247
k_T (dm^3/mg)	1043	0.16	0.61	3895
m_T	0.10	0.57	0.11	0.11
χ^2	55	17	46	70
R	0.97	0.998	0.991	0.97

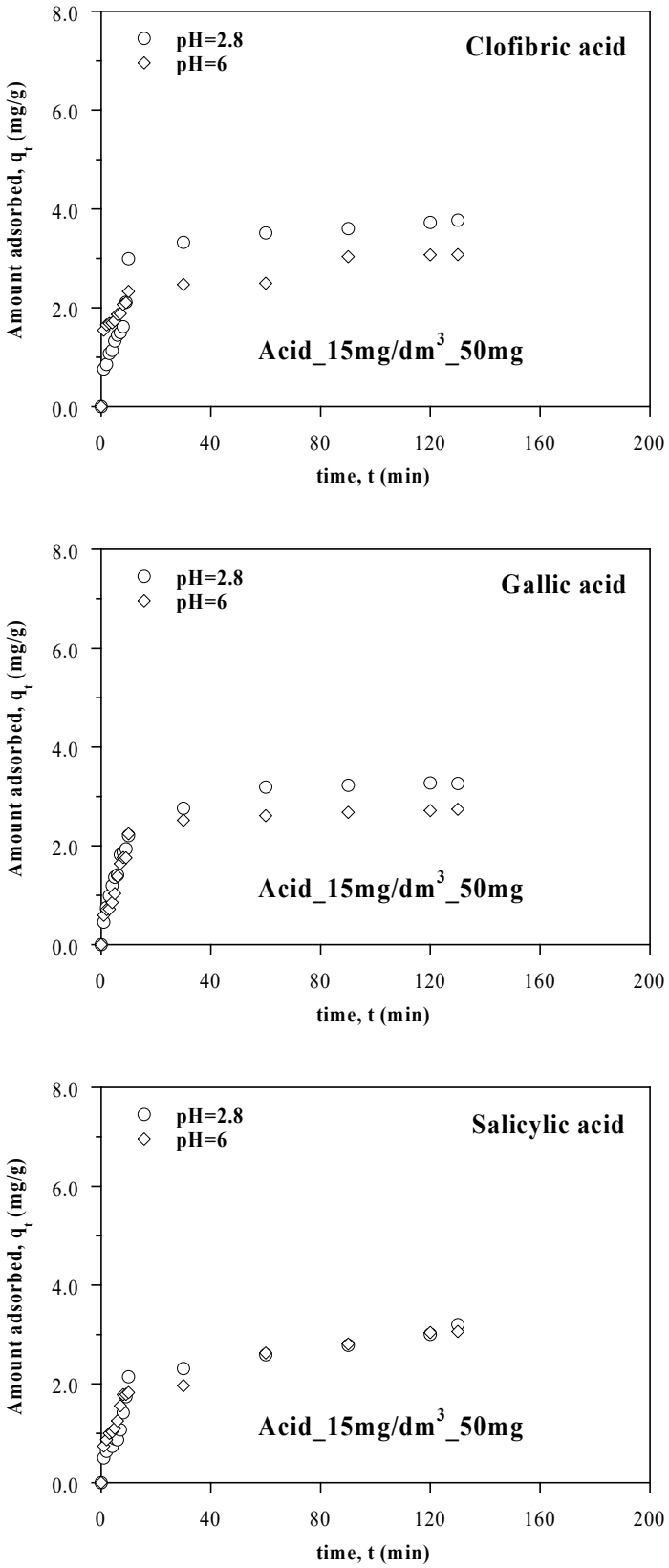


Figure 1. Kinetic adsorption data for clofibric (CA), gallic (GA) and salicylic (SA) acids on activated carbon considering two adsorption pHs (2.8 and 6). T = 298 K, 15 mg/dm^3 and 50 mg.

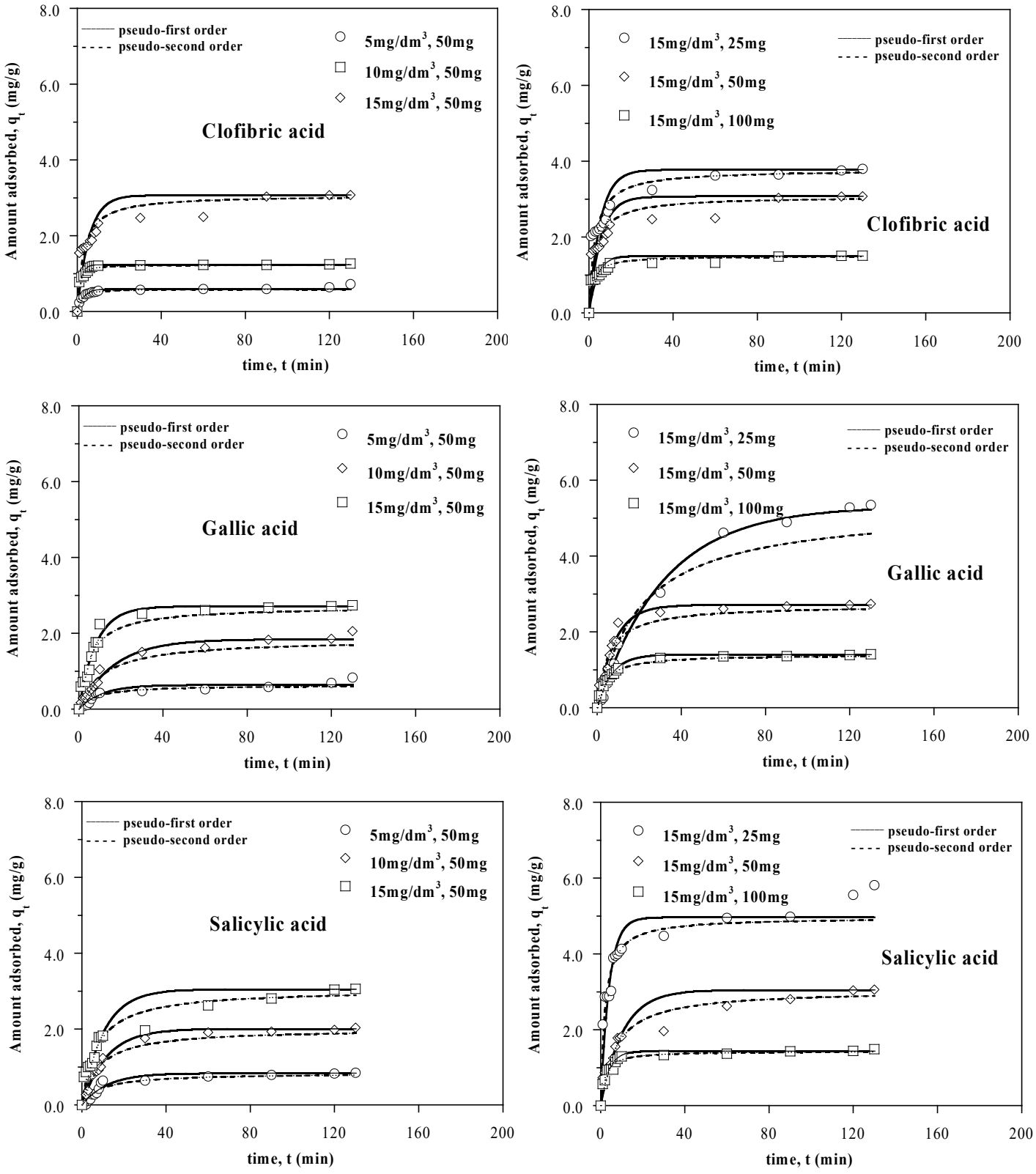


Figure 2. Kinetic adsorption data for clofibric, gallic and salicylic acids on the activated carbon considering various acid concentrations and adsorbent amounts. T = 298 K, pH 6. The lines represent a pseudo-first-order model (—) and a pseudo-second-order model (---).

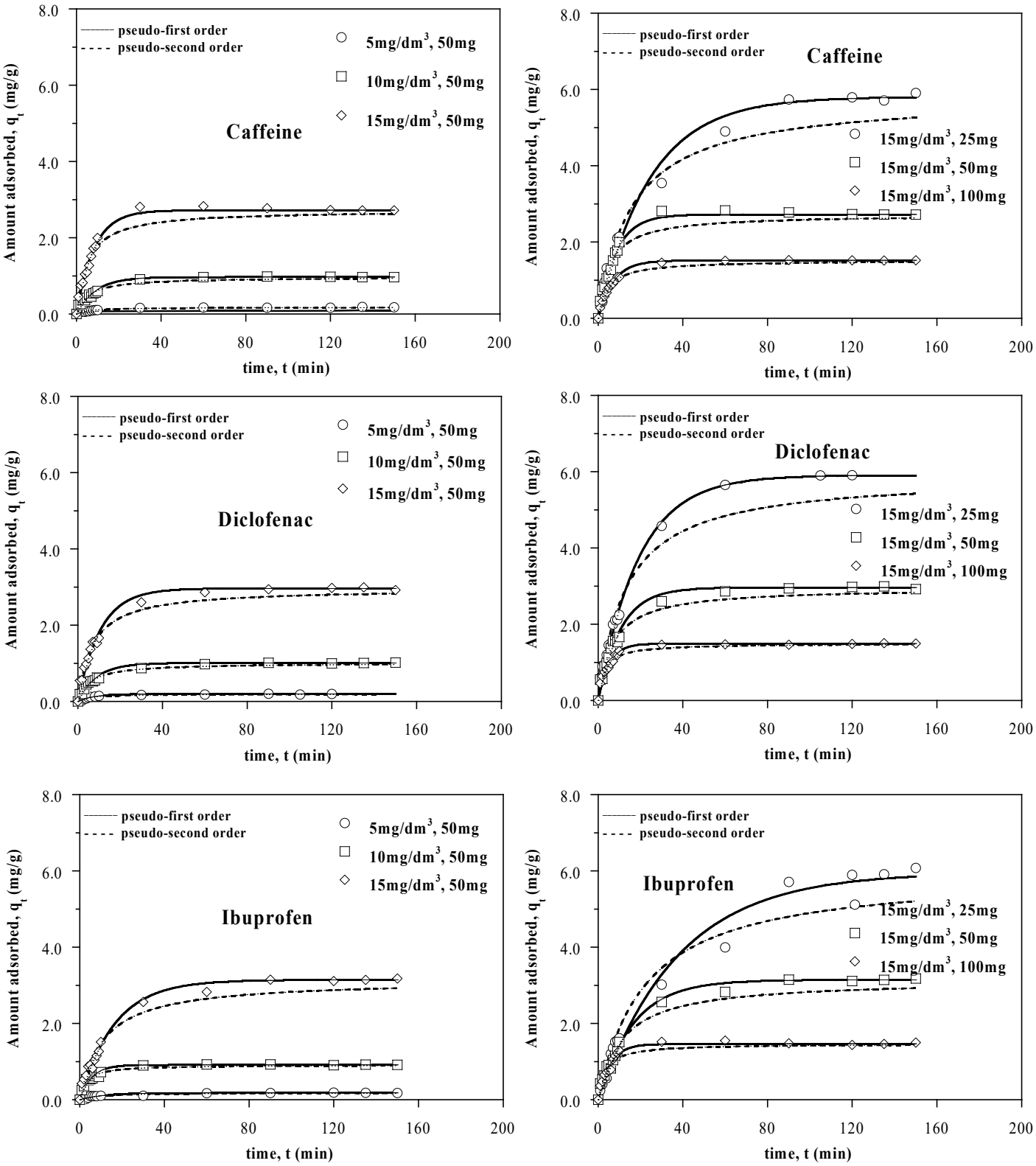


Figure 3. Kinetic adsorption data for caffeine, diclofenac and ibuprofen on the activated carbon considering various acid concentrations and adsorbent amounts. T = 298 K, pH 6. The lines represent a pseudo-first-order model (—) and a pseudo-second-order model (---).

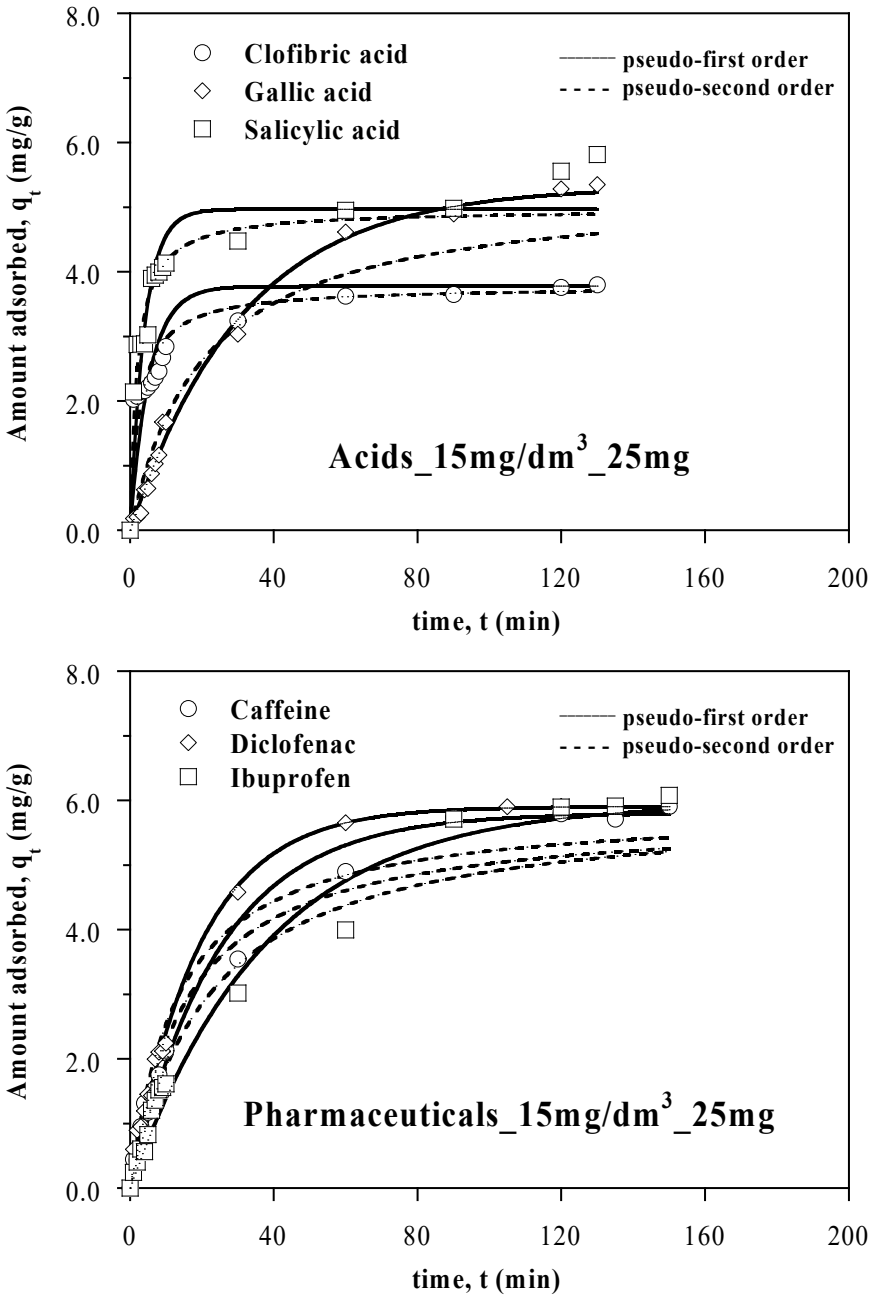


Figure 4. Kinetic adsorption data for organic molecules on the activated carbon considering 15 mg/dm^3 of acid and 25 mg of adsorbent. T = 298 K, pH 6. The lines represent a pseudo-first-order model (—) and a pseudo-second-order model (---).

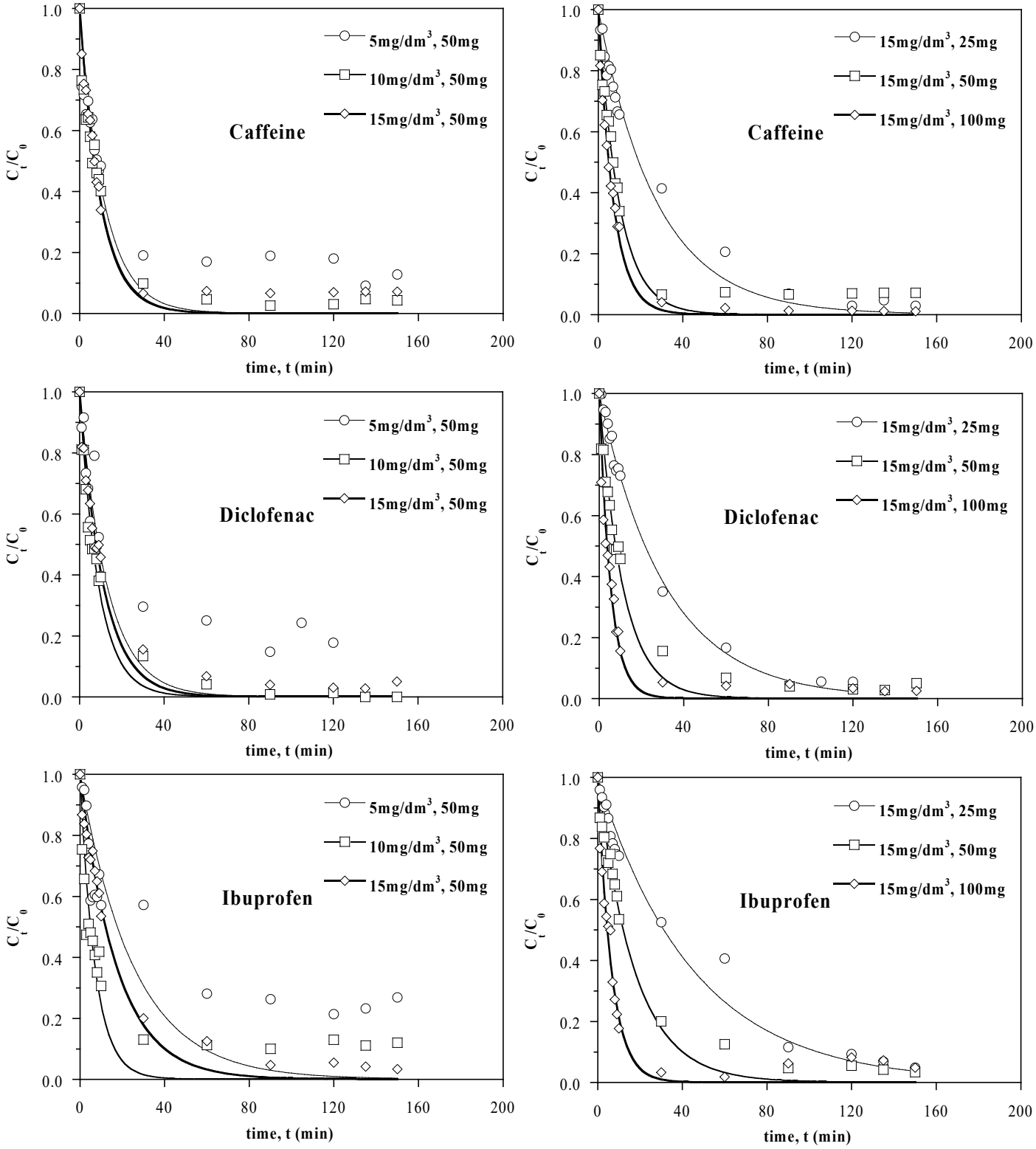


Figure 5. Furusawa–Smith model for the adsorption of organic molecules on the activated carbon. T = 298 K and pH 6.

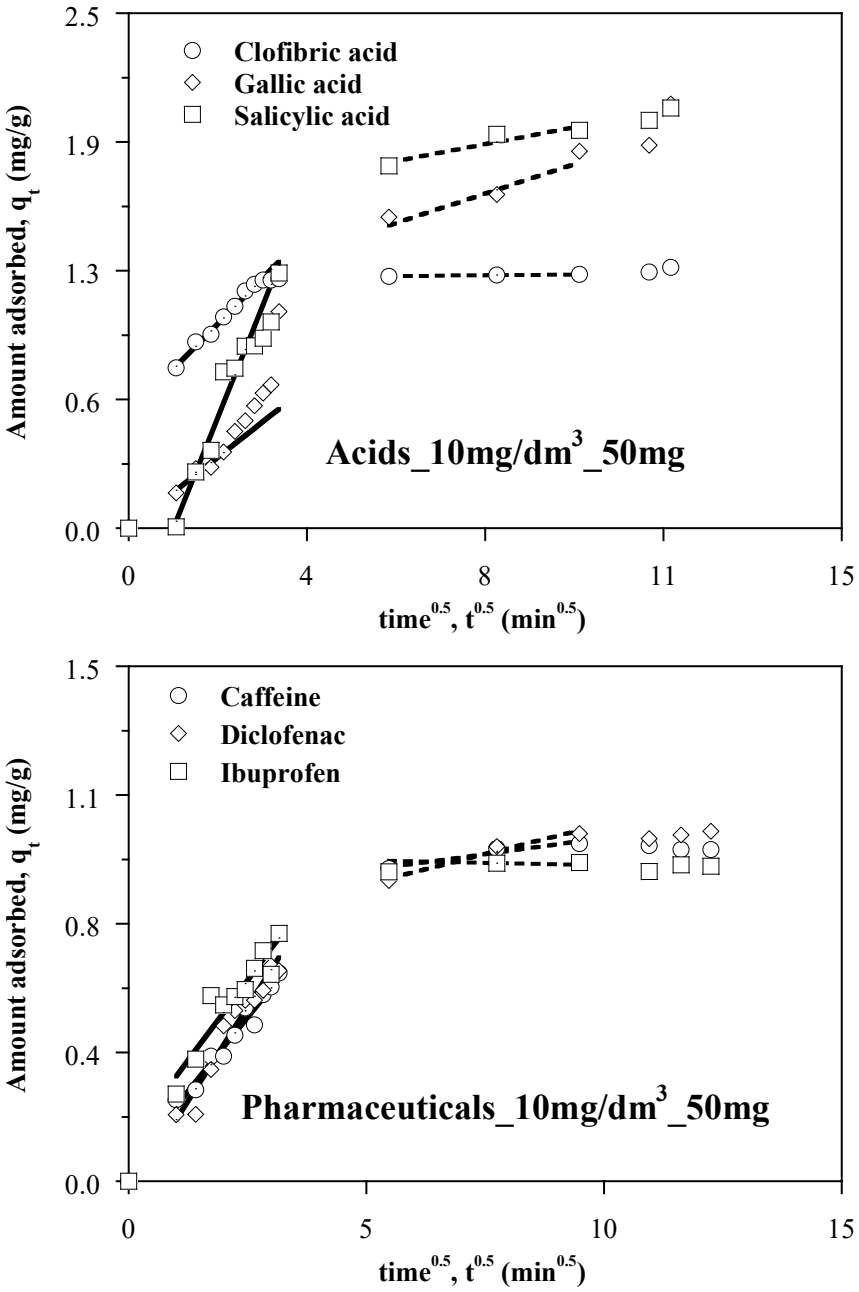


Figure 6. Intraparticle diffusion model for the adsorption of organic molecules on the activated carbon. T = 298 K and pH 6.

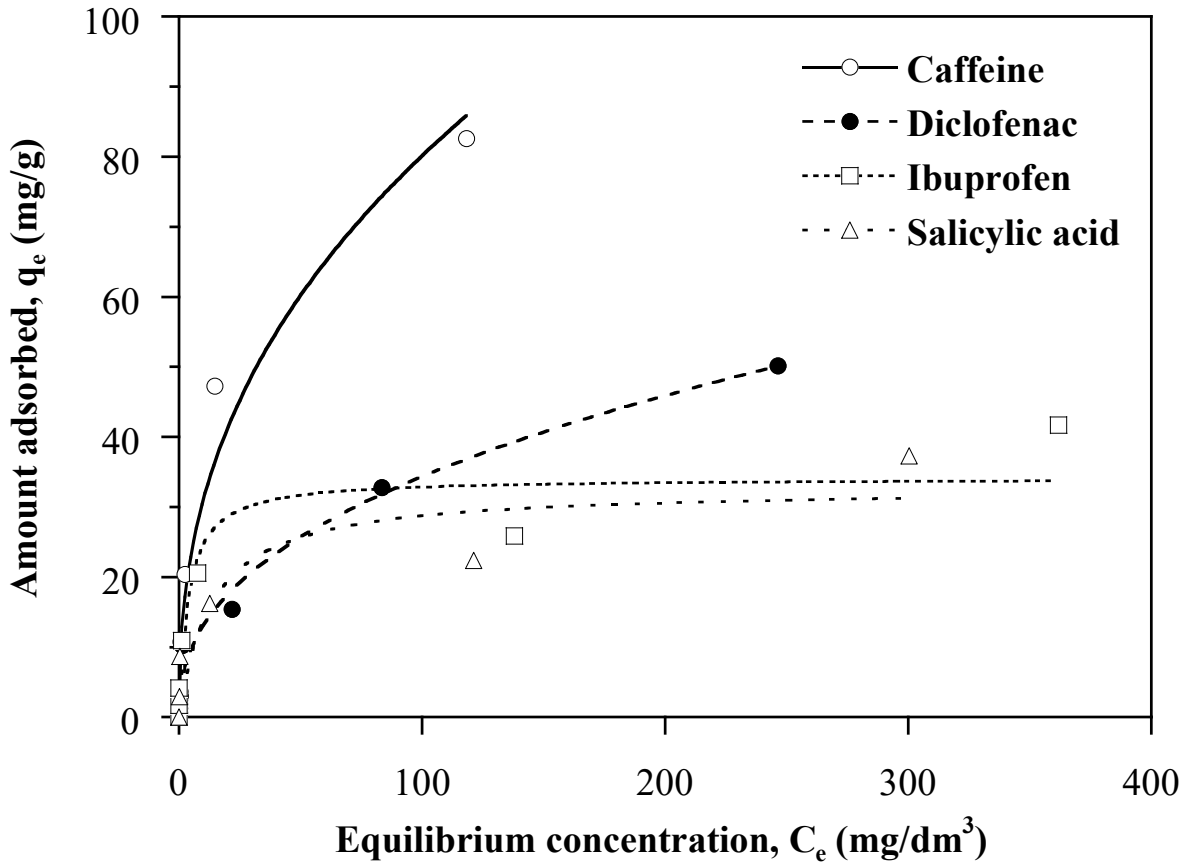


Figure 7. Experimental (symbols) and model (lines) isotherms for the equilibrium adsorption of organic molecules on the activated carbon. Equilibrium time = 2 h, T = 298 K, pH 6.